

**MASS LOSS KINETICS AND ISOTOPE FRACTIONATION OF TYPE B CAI MELTS AT 1400°C AND SOLAR  $fO_2$** ; F. M. Richter<sup>1</sup> and A. M. Davis<sup>2</sup>, <sup>1</sup>Department of the Geophysical Sciences; <sup>2</sup>Enrico Fermi Institute; University of Chicago, Chicago, IL 60637.

**Abstract.** Stolper and Paque [1] used the textures of Type B1 Ca-Al-rich refractory inclusions from carbonaceous chondrites (CAIs) to conclude that they were heated, or more likely reheated, to about 1400°C and then cooled at a rate no faster than several tens of °C per hour. CAIs are often found to have Si and Mg with isotopic compositions that are significantly fractionated, up to several ‰/amu, when compared to terrestrial values [2]. This isotopic fractionation is commonly attributed to evaporative mass loss of Si and Mg, the rate and isotopic fractionation during evaporation being interpreted with reference to the properties of evaporative residues of molten samples run at relatively high temperatures (1500–2000°C) in a vacuum furnace [3,4]. Here we present new data on the isotope fractionation and rate of mass loss of CAIB composition melts at 1400°C and 1 bar total pressure of gas with an oxygen fugacity corresponding to solar gas. We find two effects of such very reducing conditions: (1) the rate of mass loss is increased by about three orders of magnitude relative to free evaporation; and (2) there is no detectable fractionation of the Mg isotopes.

**Experimental methods.** Glass beads of REE-doped CAIB composition [5] were synthesized by suspending glass powders from preweighed Ir wire loops in a gas mixing vertical tube furnace at 1400°C and an oxygen fugacity corresponding to the iron-wustite buffer ( $\log fO_2 = -10$ ). The oxygen fugacity in this synthesis step, and in all later experiments as well, was measured using a SIRO2 yttria-stabilized zirconia oxygen fugacity probe calibrated against the IW buffer, and for more reducing conditions, against the C-CO, Cr-Cr<sub>2</sub>O<sub>3</sub>, and Ta-Ta<sub>2</sub>O<sub>5</sub> buffers [R. Mendybaev, pers. comm.]. Temperature was measured with a Pt-Rh thermocouple calibrated against the melting point of gold. The mass of glass of each bead was determined by subtracting the weight of the Ir loop from that of the loop+glass bead.

The glass beads were later returned to the gas mixing furnace and held at 1400°C for periods between 2.5 hours and 8 hours at an oxygen fugacity in the range from IW to solar  $fO_2$  (IW–6.5  $\log fO_2$  units). The samples were then reweighed to determine the rate of mass loss ( $\text{g cm}^{-2} \text{ sec}$ ), if any, as a function of  $fO_2$ . The mass loss was independently calculated from the increased concentration of REE in the final glass relative to the starting glass, and this second measure of total mass loss is in good agreement with that determined by weighing. A modified AEI IM-20

ion microprobe was used to measure the Mg isotopic composition of the starting and final glasses.

**Results.** The first important result derived from our new experiments is that the rate of mass loss from CAIB composition melts is very much dependent on  $fO_2$ , and, when conditions become very reducing (e.g., solar  $fO_2$ ) the rate of mass loss is orders of magnitude faster than the rate of free evaporation of molten samples in vacuum furnace experiments. Fig. 1 compares the mass loss rates we observed to those measured by Wang [6] in his vacuum furnace experiments on both forsterite and melts that initially had solar proportions of major rock-forming oxides. Since Wang [6] found little difference between the evaporation rate of forsterite and material of solar composition, we believe it is reasonable to compare our experiments to his despite the fact that our melts are of a different composition from those that he studied. When a comparison is made at  $T=1400^\circ\text{C}$ , we find that mass loss at an  $fO_2$  of IW–5 is more than two orders of magnitude faster than that in the vacuum experiments and that by IW–6.5 (solar  $fO_2$ ) the rate is about three orders of magnitude faster. Given the low temperature and short duration of our experiments we did not measure any significant mass loss for conditions more oxidizing than IW–5.

Fig. 2 shows what we believe is an equally important, and somewhat surprising, result: despite substantial mass loss of Mg from our samples run at solar  $fO_2$ , the Mg isotopic composition of the residues is identical to that of the starting material. For comparison, Fig. 2 shows the Mg isotope fractionation of molten forsterite residues evaporated under vacuum [3]. One possible explanation of our not having found any Mg isotope fractionation is that the diffusion of Mg was sufficiently slow to inhibit the isotopic fractionation of the bulk sample. This is not the case in our experiments because we found no measurable chemical gradients in Mg.

**Discussion.** The results presented above are limited in their applicability to the interpretation of actual CAIs by the fact that, while we have carried out experiments over a range of oxygen fugacities, the total pressure, and thus the partial pressure of hydrogen, is many orders of magnitude greater than that of the environment in which they are thought to have CAIs formed. Furthermore, when we compare our results to what has been found in vacuum experiments, we need to keep in mind that we are comparing experiments on materials of different chemical composition.

Despite the preliminary nature of our results we

## MASS LOSS AND ISOTOPIC FRACTIONATION OF MOLTEN CAI MELT; Richter and Davis

are excited by the prospect that we may be on our way to developing a cosmobarometer that will enable us to place an upper bound on the pressure of the environment in which CAIs formed. We can illustrate our idea for a cosmobarometer by making use of Nagahara and Ozawa's [7] data for the mass loss kinetics of forsterite as a function of the partial pressure of hydrogen in the range  $10^{-9}$  to  $10^{-4}$  bars. Fig. 3 shows a curve for mass loss kinetics that falls through Nagahara and Ozawa's [7] data after their data was adjusted from  $T=1700^{\circ}\text{C}$  at which their experiments were run to  $T=1400^{\circ}\text{C}$  of our experiments using the activation energy for evaporation from [6]. The comparison is made at  $T=1400^{\circ}\text{C}$  because this is a reasonable peak temperature for Type B CAIs [1]. At sufficiently low pressure ( $P_{\text{H}_2} < 10^{-6}$  bars) evaporation kinetics are independent of pressure and would seem to correspond to free evaporation, which is the regime in which isotope fractionation is known to occur. The increase in the rate of mass loss for  $P_{\text{H}_2} > 10^{-6}$  is most likely due to surface reactions with hydrogen. Once hydrogen reactions become the dominant mechanism for mass loss, the kinetics become faster, and we suggest on the basis of our solar  $f\text{O}_2$  experiments that there will no longer be any significant isotope fractionation associated with the loss of elements such as Mg and Si. Thus, one can imagine there being two regimes, a free evaporation-dominated regime with slow mass loss kinetics and isotope fractionation; and a hydrogen reaction-dominated regime with faster mass loss kinetics but little or no measurable isotope fractionation. Figure 3 suggests that the transition between these two regimes is in the pressure range  $P_{\text{H}_2} = 10^{-6}$ – $10^{-5}$  bars, implying that CAIs with measurable and correlated isotopic fractionations of Mg and Si must have formed in an environment with  $P_{\text{H}_2} < 10^{-5}$  bar in order to have been in the isotope fractionating free-evaporation dominated regime.

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